

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

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COMMISSIONER FOR PATENTS Washington, D.C. 20231

Attorney Docket No.: <u>DAIN:447A</u>
Date: <u>October 6, 2000</u>

Prior Application:

Examiner: C. H. Kelly

Art Unit: 1756

Sir:

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This is a request for filing a

[] Continuation [X] Divisional (parent not abandoned) application under 37 C.F.R. §1.53(b), of pending prior application Serial No. 09/025,249, filed February 18, 1998

for [Title]: LIQUID CRYSTALLINE COMPOUNDS AND PROCESS FOR PRODUCING THE SAME by [Inventors]: Junichi HANNA, Masahiro FUNAHASHI, Komei KAFUKU, and

Kyoko KOGO

- [X] A copy of the prior application is attached. This copy comprises a true copy of the prior application as filed (specification, claims, drawings, declaration). No amendments referred to in the declaration (if any) filed to complete the prior application introduced new matter therein.
- [X] The filing fee is calculated below:

CLAIMS AS FILED IN THE PRIOR APPLICATION, LESS ANY CLAIMS
CANCELLED BY AMENDMENT BELOW
Basic Fee

\$710.00 (*35.00)

Total claims 19 -20 = 0 x \$18.00 (* 9.00) =
Independent claims 1 -3 = 0 x \$80.00 (*40.00) = $\frac{-}{$710.00}$

- 3. [X] A check in the amount of \$710.00 is enclosed (Ck# 13239).

 THE COMMISSIONER IS HEREBY AUTHORIZED TO CHARGE ANY OTHER FEES WHICH MAY BE REQUIRED OR CREDIT ANY OVERPAYMENT TO DEPOSIT ACCOUNT NO. 16-0331. TWO DUPLICATE COPIES OF THIS FORM ARE ENCLOSED.
- 4. [] Cancel in this application original claims ____ of the prior application before calculating the filing fee. At least one original independent claim is retained for filing purposes.
- 5. [X] Amend the specification by inserting before the first line the sentence: --This is a [] Continuation [X] Division of application Serial No. 09/025,249 filed February 18, 1998.--

PLEASE ACCEPT THIS AS AUTHORIZATION TO DEBIT OR CREDIT FEES TO DEP. ACCT. 16-0331 PARKHURST & WENDE!

- 6. [] Transfer the drawings from the prior application to this application and abandon said prior application as of the filing date accorded this application. A duplicate copy of this form is enclosed for filing in the prior application file.
- 7. [] New [] Formal [] Informal drawings are enclosed: Fig(s). .
- 8. [X] Priority of foreign application(s) No. 49593/1997 filed February 19, 1997 in Japan is claimed under 35 U.S.C. §119.
 - [X] The certified copy was filed in prior application No. 09/025,249 on April 3, 1998.
 - [] A certified copy of the above corresponding foreign application is filed herewith.
- 9 [X] The prior application is assigned of record to <u>DAI NIPPON PRINTING</u> <u>CO., LTD.</u> Recorded at Reel <u>9115</u>, Frame <u>0068</u>.
- 10. [X] The power of attorney in the prior application is to Roger W. Parkhurst, Registration No. 25,177 and Charles A. Wendel, Registration No. 24,453:
 - [X] a. The power appears in the original papers in the prior application.
 - [] b. Since the power does not appear in the original papers, a copy of the power in the prior application is enclosed.
 - [X] c. Address all future communications to

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PARKHURST & WENDEL, L.L.P. 1421 Prince Street, Suite 210 Alexandria, Virginia 22314-2805 Telephone: (703) 739-0220

11. [X] A Preliminary Amendment is enclosed. Claims added by this Amendment are properly numbered consecutively beginning with the number next following the highest numbered original claim in the prior application.

- 12.
- [X] An Information Disclosure Statement is enclosed.
- *13. [] Small entity status was established in the parent application via a verified statement filed on _______.

Respectfully submitted,

PARKHURST & WENDEL, L.L.P.

Charles A. Wendel

Registration No. 24,453

(**Fev**. 3/99)

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of

Junichi HANNA et al.

Prior

Group Art Unit: 1756

Serial No.: Rule 53(b) Div. of

Serial No. 09/025,249

Filed: February 18, 1998

Prior

Filed: October 6, 2000

Examiner: C. H. Kelly

For: LIQUID CRYSTALLINE COMPOUNDS AND PROCESS FOR

PRODUCING THE SAME

PRELIMINARY AMENDMENT

Commissioner for Patents Washington, D.C. 20231

Sir:

Prior to examination of the above-identified application, please enter the following specification and claim changes as noted below:

IN THE SPECIFICATION:

Page 1, line 11, insert --an-- after "using".

IN THE CLAIMS:

Cancel claims 1, 3, and 7 without prejudice or disclaimer.

Claim 5, line 1, cancel "1 or".

Claim 9, line 1, cancel "7 or".

Claims 11 and 12, both line 2, cancel "1 or".

Claim 13, line 1, cancel "1 or".

Claim 14, line 2, cancel "1 or".

Claim 15, line 1, cancel "1"; and

line 2, cancel "or".

Claim 16, line 1, cancel "1 or".

Claims 17 and 18, both line 2, cancel "or 6".

Claim 19, line 1, cancel "or"; and

line 2, cancel "6".

Claims 20 and 21, both line 2, cancel "or 6".

Claim 22, line 1, cancel "or 6".

REMARKS

This application is a Rule 53(b) divisional application of U.S. Serial No. 09/025,249, filed February 18, 1998, now allowed.

Claims 1, 3, and 7 have been canceled and multiple claim dependencies have been eliminated where appropriate. The claims presented for examination are claims 2, 4 to 6, and 8 to 22.

The specification has been amended in the same manner as in the parent application and to identify the parent application.

Filed herewith is an Information Disclosure Statement listing all references cited during prosecution of the parent application.

Prompt examination of this application on the merits is respectfully solicited.

Respectfully submitted,

PARKHURST & WENDEL, L.L.P.

Charles A. Wendel

Registration No. 24,453

CAW/ch

Attorney Docket No. DAIN: 447A PARKHURST & WENDEL, L.L.P. 1421 Prince Street, Suite 210 Alexandria, Virginia 22314-2805 Telephone: (703) 739-0220

(rev. 3/3/00)

LIQUID CRYSTALLINE COMPOUNDS AND PROCESS FOR PRODUCING THE SAME

BACKGROUND OF THE INVENTION

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The present invention relates to liquid crystalline compounds and more particularly to novel crystalline compounds, which exhibit liquid crystallinity and, in addition, charge transport capability, and a process for producing the same.

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Liquid crystalline compounds having various structures are known in the art and are widely used mainly as materials for information display devices using electro-optic effect based on the alignment effect of liquid crystal molecules attained by application of voltage. Further, application of liquid crystalline compounds to optical shutters, optical stops, modulating devices, lenses, light beam deflection/optical switches, phase diffraction gratings, optical logic devices, memory devices and the like are under study. External stimulation by heat, electric field, magnetic field, pressure or the like results in transition of the alignment of liquid crystal molecules which enables optical properties and electric capacity to be easily changed. Sensors and measuring instruments, utilizing this property, for temperature, electric field/voltage, infrared radiation, ultrasonic wave, flow rate/acceleration, gas or pressure have been studied in the art.

DISCLOSURE OF THE INVENTION

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An object of the present invention is to provide liquid crystalline compounds having a novel structure and a process for producing the same.

The above object can be attained by the following present invention. Specifically, according to one aspect of the present invention, there is provided a liquid crystalline compound represented by the following general formula (I):

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wherein R_1 and R_2 each independently represent a straight-chain, branched or cyclic, saturated or unsaturated hydrocarbon group having 1 to 22 carbon atoms and may be attached directly to the aromatic ring without through X_1 or X_2 ; R_3 represents a hydrogen atom, a cyano group, a nitro group, a fluorine atom, or a methyl group; and X_1 and X_2 each independently represent an oxygen atom, a sulfur atom, or a -CO-, -OCO-, -COO-, -N=CH-, -CONH-, -NH-, -NHCO-, or -CH₂- group.

According to another aspect of the present invention, there is provided a liquid crystalline compound represented by the following general formula (II):

wherein R_1 and R_2 each independently represent a straight-chain, branched or cyclic, saturated or unsaturated hydrocarbon group having 1 to 22 carbon atoms and may be attached directly to the aromatic ring without through X_1 or X_2 ; R_3 represents a hydrogen atom, a cyano group, a nitro group, a fluorine atom, or a methyl group; X_1 and X_2 each independently represent an oxygen atom, a sulfur atom, or a -CO-, -OCO-, -COO-, -N=CH-, -CONH-, -NH-, -NHCO-, or -CH₂- group; and Z represents a -COO-, -OCO-, -N=N-, -CH=N-, -CH₂S-, -CH=CH-, or -C = C- group.

According to still another aspect of the present invention, there is provided a process for producing the liquid crystalline compound represented by the general formula (I), comprising the step of reacting a compound represented by the following general formula (1) with a compound represented by the following general formula (2):

R₁-X₁—
$$B(OH)_2$$
 (1)

$$Br \longrightarrow X_2-R_2$$
 (2)

wherein R_1 , R_2 , R_3 , X_1 , and X_2 are as defined above.

According to a further aspect of the present invention, there is provided a process for producing the liquid crystalline compound represented by the general formula (II), comprising the step of reacting a compound represented by the following general formula (3) with a compound represented by the following general formula (4):

$$R_1-X_1 \longrightarrow Y_1 \tag{3}$$

$$Y_2 \longrightarrow X_2-R_2 \qquad (4)$$

wherein R_1 , R_2 , R_3 , X_1 , and X_2 are as defined above; and Y_1 and Y_2 are respectively groups which are reacted with each other to form a -COO-, -OCO-, -N=N-, -CH=N-, -CH₂S-, -CH=CH-, or -C \equiv C- group.

The present invention can provide novel liquid crystalline compounds having not only liquid crystallinity but also charge transport capability. The novel liquid crystalline compounds can be used in applications, where the conventional liquid crystalline compounds are used, and, in addition, are useful as materials for optical sensors, electroluminescence devices,

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photoconductors, space light modulating devices, thin film transistors, other sensors and the like, utilizing the charge transfer capability. In particular, some of the liquid crystalline compounds of the present invention have both electron transport capability and hole transport capability and, when mixed with a fluorescent material in order to use them as a material for an electroluminescence device, can provide luminescence.

The present invention will be described in more detail with reference to the following preferred embodiments.

Example 1

50 ml of THF (tetrahydrofuran) was added to 2.91 g (0.12 mol) of metallic magnesium, and the mixture was stirred. 100 ml of a solution of 26.89 g (0.1 mol) of p-octylbromobenzene in THF was added dropwise thereto, and the mixture was heated. After the initiation of the reaction was confirmed, the mixture was refluxed for one hr. The mixture was cooled to -78°C, 12.46 g (0.12 mol) of trimethylboric acid was added dropwise thereto, and the mixture was stirred for 30 min. The temperature was returned to room temperature, followed by stirring for additional one hr. Dilute hydrochloric acid was added thereto, and the mixture was stirred for one hr. The aqueous layer was extracted with ether, and the oil layer was washed with water and then with an aqueous sodium hydrogencarbonate. The oil layer was then dried over sodium sulfate, the solvent was removed by distillation, and the resultant crude product was purified by chromatography on silica gel to give p-octylphenylboric acid.

The above compound exhibited the following peaks in NMR spectrum:

¹H NMR (CDCl₃)

 $\delta = 8.14 \; (2\text{H, d, J} = 8.6 \; \text{Hz}), \, 7.31 \; (2\text{H, d, J} = 7.9 \; \text{Hz}), \, 2.68 \; (2\text{H, t, J} = 7.3 \; \text{Hz}), \, 1.50\text{-}1.80 \; (4\text{H, m}), \, 1.20\text{-}1.40 \; (8\text{H, m}), \, 0.88 \; (3\text{H, t, J} = 7.6 \; \text{Hz})$

22.29 g (0.1 mol) of 2-bromo-6-naphthol, 11.22 g (0.2 mol) of potassium hydroxide, and 32.36 g (0.13 mol) of 1-bromododecane were dissolved in ethanol (300 ml), and the solution was refluxed for 8 hr. Thereafter, water was added thereto, the mixture was cooled, and the resultant precipitate was collected by filtration and washed with a sodium hydroxide solution and then with water. The crude product thus obtained was recrystallized from ethyl acetate to give 2-bromo-6-dodecyloxynaphthalene.

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The above compound exhibited the following peaks in NMR spectrum:

¹H NMR (CDCl₂)

 $\delta = 7.89 \ (1\text{H, d, J} = 2.0 \ \text{Hz}), \ 7.62 \ (1\text{H, d, J} = 8.9 \ \text{Hz}), \ 7.57 \ (1\text{H, d, J} = 8.9 \ \text{Hz}), \ 7.47 \ (1\text{H, dd, J1} = 2.0 \ \text{Hz}, \ \text{J2} = 8.9 \ \text{Hz}), \ 7.15 \ (1\text{H, dd, J1} = 2.6 \ \text{Hz}, \ \text{J2} = 8.9 \ \text{Hz}), \ 7.07 \ (1\text{H, d, J} = 2.6 \ \text{Hz}), \ 4.04 \ (2\text{H, t, J} = 6.6 \ \text{Hz}), \ 1.84 \ (2\text{H, quint, J} = 6.6 \ \text{Hz}), \ 1.40-1.50 \ (4\text{H, m}), \ 1.17-1.40 \ (14\text{H, m}), \ 0.88 \ (3\text{H, t, J} = 6.8 \ \text{Hz})$

2.01~g~(0.01~mol) of p-octylphenylboric acid, 3.91~g~(0.01~mol) of 2-bromo-6-dodecyloxynaphthalene, and $Pd(PPh_3)_4~(0.0005~mol)$ were dissolved in dimethoxyethane (50 ml), a 10% aqueous potassium carbonate solution (40 ml) was added thereto, and the mixture was refluxed for one hr. After cooling, the resultant precipitate was collected by filtration and washed with water and ethanol. The crude product thus obtained was recrystallized from hexane to give a compound represented by the following formula:

The above compound exhibited the following peaks in NMR spectrum: ¹H NMR (CDCl₃)

 δ = 7.94 (1H, d, J = 1.3 Hz), 7.77 (2H, d, J = 8.6 Hz), 7.69 (1H, dd, J1 = 1.7 Hz, J2 = 8.6 Hz), 7.62 (2H, d, J = 8.3 Hz), 7.28 (2H, d, J = 8.3 Hz), 7.16 (1H, dd, J1 = 2.6 Hz, J2 = 8.3 Hz), 7.14 (1H, s), 4.08 (2H, t, J = 6.6 Hz), 2.66 (2H, t, J = 7.3 Hz), 1.86 (2H, quint, J = 6.8 Hz), 1.40-1.70 (4H, m), 1.20-1.70 (26H, m), 0.89 (3H, t, J = 5.6 Hz), 0.88 (3H, t, J = 6.9 Hz)

The above compound had the following phase transition temperatures.

Crystal - 79.3°C - SmX₁ - 100.4°C - SmX₂ - 121.3°C - Iso. (X₁ and X₂ were unidentified)

The charge mobility of the above compound was 10⁻³ cm²/Vs for both electron and hole.

30 Example 2

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2.18 g (0.01 mol) of p-octylbenzaldehyde and 3.28 g (0.01 mol) of 2-amino-6-dodecyloxynaphthalene were dissolved in ethanol (30 ml), and the solution was heated at 70°C for 2 hr with stirring. After the reaction, the mixture was cooled to room temperature, and the precipitated solid was collected by filtration and recrystallized from ethanol to give a compound represented by the following formula. This compound had the same properties as the compound prepared in Example 1.

Example 3

The procedure of Example 1 was repeated to prepare liquid crystalline compounds represented by the general formula (I) wherein R_1 , R_2 , R_3 , X_1 , and X_2 represent respective groups specified in Table 1. All the liquid crystalline compounds thus obtained had the same properties as the liquid crystalline compound prepared in Example 1.

Table 1

Table I							
R ₁	R ₂	R_3	X ₁	X ₂			
CH ₃ (CH ₂) ₈	(CH ₂) ₉ CH ₃	Н	CH,	0			
CH ₃ (CH ₂) ₅	(CH ₂) ₇ CH ₃	3'-CN	CH ₂	S			
CH ₃ (CH ₂) ₁₅	(CH ₂) ₃ CH ₃	2'-F	0	0			
CH ₃ CH ₂ C*H(CH ₃)CH	(CH ₂) ₅ CH ₃	3'-NO ₂	S	0			
CH ₃ (CH ₂) ₈	CH ₃ (CH ₂) ₈	Н	CH ₂	CH ₂			
C ₅ H ₁₁ CFCH ₃	C ₁₀ H ₂₁	Н	C00	0			
C ₈ H ₁₇	C₅H₁₁	2'-F, 3'- F	0	-			
	CH ₃ (CH ₂) ₈ CH ₃ (CH ₂) ₅ CH ₃ (CH ₂) ₁₅ CH ₃ CH ₂ C*H(CH ₃)CH 2 CH ₃ (CH ₂) ₈ C ₅ H ₁₁ CFCH ₃	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$			

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Example 4

The procedure of Example 2 was repeated to prepare liquid crystalline compounds represented by the general formula (II) wherein R_1 , R_2 , R_3 , X_1 , X_2 , and Z represent respective groups specified in Table 2. All the liquid crystalline compounds thus obtained had the same properties as the liquid crystalline compound prepared in Example 2.

Table 2

Ex.	R,	R,	R ₃	Χ,	Х,	Z
4-1	CH ₃ (CH ₂) ₈	(CH ₂) ₉ CH ₃	2',3'-F	CH,	0	CH=N
4-2	CH ₃ (CH ₂) ₅	(CH ₂) ₇ CH ₃	3'-CN	CH ₂	S	CO0
4-3	CH ₃ CH ₂ C*H(CH ₃)CH ₂	(CH ₂) ₅ CH ₃	3'-NO ₂	CH₂	0	CH=C
E c						Н
4-4	CH ₃ (CH ₂) ₁₅	(CH ₂) ₃ CH ₃	Н	0	0	C≡C
4-5	CH ₃ (CH ₂) ₈	CH ₃ (CH ₂) ₈	Н	CH,	CH ₂	N=N
4-6	C₄H ₉	C ₆ H ₅ -C₄H ₉	Н	-	co	oco
					0	
4-7	C ₂ H ₅ CH(CH ₃)CH ₂	C ₁₀ H ₂₁	H	000	0	CO
4-8	C ₆ H ₁₃ OC ₆ H ₅	Н	Н	CH=	-	CH=N
				N		

Example 5

Two glass substrates each having an ITO electrode (surface electric resistance: 100-200 Ω/\Box) provided by vacuum film formation were laminated onto each other so that the ITO electrodes faced each other while providing a gap (about $2 \mu m$) therebetween using spacer particles, thereby preparing a cell. (2-(4'-octylphenyl)-6-The naphthalene compound liquid crystal dodecyloxynaphthalene, Crystal - 79°C - SmX - 121°C - Iso.) prepared in Example 1 was mixed with 1% by mole of a luminescent dye (3-(2benzothiazolyl)-7-(diethylamino)-2H-1-benzopyran-2-one (manufactured by Nihon Kanko Shikiso Kenkyusho (Japan Photosensitive Dye Laboratory), oscillating wavelength: 507-585 nm), and the mixture was poured at 125 $^{\circ}\text{C}$ into the cell. An d.c. electric field of 250 V was applied to the cell in a dark place.

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As a result, light emission derived from the fluorescent wavelength of the fluorescent dye was observed.

As described above, the present invention can provide novel liquid crystalline compounds having not only liquid crystallinity but also charge transport capability. The novel liquid crystalline compounds can be used in applications, where the conventional liquid crystalline compounds are used, and, in addition, are useful as materials for optical sensors, electroluminescence devices, photoconductors, space light modulating device, thin film transistors, other sensors and the like, utilizing the charge transfer capability. In particular, some of the liquid crystalline compounds of the present invention have both electron transport capability and hole transport capability and, when mixed with a fluorescent material in order to use it as a material for an electroluminescence device, can provide luminescence.

CLAIMS

1. A liquid crystalline compound represented by the following general formula (I):

$$R_1-X_1$$
 R_2 R_3 R_1-X_1 R_2 R_3

wherein R_1 and R_2 each independently represent a straight-chain, branched or cyclic, saturated or unsaturated hydrocarbon group having 1 to 22 carbon atoms and may be attached directly to the aromatic ring without through X_1 or X_2 ; R_3 represents a hydrogen atom, a cyano group, a nitro group, a fluorine atom, or a methyl group; and X_1 and X_2 each independently represent an oxygen atom, a sulfur atom, or a -CO-, -OCO-, -COO-, -N=CH-, -CONH-, -NH-, -NHCO-, or -CH₂- group.

2. A liquid crystalline compound represented by the following general formula (II):

wherein R_1 and R_2 each independently represent a straight-chain, branched or cyclic, saturated or unsaturated hydrocarbon group having 1 to 22 carbon atoms and may be attached directly to the aromatic ring without through X_1 or X_2 ; R_3 represents a hydrogen atom, a cyano group, a nitro group, a fluorine atom, or a methyl group; X_1 and X_2 each independently represent an oxygen atom, a sulfur atom, or a -CO-, -OCO-, -COO-, -N=CH-, -CONH-, -NH-, -NHCO-, or -CH₂- group; and Z represents a -COO-, -OCO-, -N=N-, -CH=N-, -CH₂S-, -CH=CH-, or -C \equiv C- group.

3. A process for producing the liquid crystalline compound according to

claim 1, comprising the step of reacting a compound represented by the following general formula (1) with a compound represented by the following general formula (2):

$$R_3$$
 R_1-X_1
 \longrightarrow
 $B(OH)_2$ (1)

wherein R_1 , R_2 , R_3 , X_1 , and X_2 are as defined above.

4. A process for producing the liquid crystalline compound according to claim 2, comprising the step of reacting a compound represented by the following general formula (3) with a compound represented by the following general formula (4):

$$R_1-X_1 \longrightarrow Y_1$$
 (3)

$$Y_2 \longrightarrow X_2-R_2 \qquad (4)$$

wherein R_1 , R_2 , R_3 , X_1 , and X_2 are as defined above; and Y_1 and Y_2 are respectively groups which are reacted with each other to form a -COO-, -OCO-, -N=N-, -CH=N-, -CH₂S-, -CH=CH-, or -C \equiv C- group.

5. The liquid crystalline compound according to claim 1 or 2, which has charge transport capability.

- 6. The liquid crystalline compound according to claim 5, which has a liquid crystal phase comprising at least a smectic phase.
- 7. The liquid crystalline compound according to claim 1, wherein R_3 represents a hydrogen or fluorine atom and X_1 and X_2 each independently represent an oxygen atom or a -CH₂-, -CO-, -COO-, or -N=CH-group.
- 8. The liquid crystalline compound according to claim 2, wherein R_3 represents a hydrogen or fluorine atom and X_1 and X_2 each independently represent an oxygen atom or a -CH₂-, -CO-, -COO-, or -N=CH-group.
- 9. The liquid crystalline compound according to claim 7 or 8, which has charge transport capability.
- 10. The liquid crystalline compound according to claim 9, which has a liquid crystal phase comprising at least a smectic phase.
- 11. An image display device comprising the compound according to claim 1 or 2 in a drive path.
- 12. An electroluminescence device comprising the compound according to claim 1 or 2 in a drive path.
- 13. A photoconductor comprising the compound according to claim 1 or 2 in a drive path.
- 14. A space light modulating device comprising the compound according to claim 1 or 2 in a drive path.
- 15. A thin film transistor comprising the compound according to claim 1 or 2 in a drive path.
- 16. A sensor comprising the compound according to claim 1 or 2 in a drive path.
- 17. An image display device comprising the compound according to claim 5 or 6 in a drive path.
- 18. An electroluminescence device comprising the compound according to claim 5 or 6 in a drive path.
- 19. A photoconductor comprising the compound according to claim 5 or 6 in a drive path.
 - 20. A space light modulating device comprising the compound according

to claim 5 or 6 in a drive path.

- 21. A thin film transistor comprising the compound according to claim 5 or 6 in a drive path.
- 22. A sensor comprising the compound according to claim 5 or 6 in a drive path.

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ABSTRACT

A liquid crystalline compound having a novel structure and a process for producing the same are provided. The liquid crystalline compound is represented by the following general formula (I):

$$R_1-X_1 \longrightarrow X_2-R_2 \qquad (I)$$

wherein R_1 and R_2 each independently represent a straight-chain, branched or cyclic, saturated or unsaturated hydrocarbon group having 1 to 22 carbon atoms and may be attached directly to the aromatic ring without through X_1 or X_2 ; R_3 represents a hydrogen atom, a cyano group, a nitro group, a fluorine atom, or a methyl group; and X_1 and X_2 each independently represent an oxygen atom, a sulfur atom, or a -CO-, -OCO-, -COO-, -N=CH-, -CONH-, -NH-, -NHCO-, or -CH₂- group.

APPLICATION FOR UNITED STATES PATTITE Declaration and Power of Attorney

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name; that

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if alural inventors as	re named below)	of the subject m	atter which is clair	ned and for which	isted below) or an original, first and j a patent is sought on the invention ent OUCING THE SAME	
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*a. [] attac						
b. [X] filed	on <u>February</u>	7 18,1998as Ai	pplication Serial No	o. <u>09/025,249</u>	and amended on(if applicable)	ن
					(if applicable)	
I hereby sta			rstand the contents	of the above-ident	fied application, including the claims	, as amended
	e 37, Code of Fe	ederal Regulations	s, §1.56(a). Under	Title 35 U.S. Code	material to the examination of this a §119, the priority benefits of the follo	
Japanese	Patent Ap	pplication	No. 49593	/1997 file	d on February 19, 199	7
					re filed in countries foreign to the Un of the above-named foreign priority a	
imencan etther (a) i	more than one ye	cat prior to this a	pplication, or (b) b	tione the filling trace	of the above-named foreign priority a	ippiication(s).
2 If there are no cor insert "NONE".	responding appl	lications,				
hereby ap o transact all busine	•		ys of record with fu	ll power of substitu	tion and revocation to prosecute this a	pplication and
is						
Roger W. I	Parkhurst, Reg. 1	No. 25,177; Charl	les A. Wendel, Re	g. No. 24,453; and/	or Lawrence D. Eisen, Reg. No. 41,00)9
	DDECDAN	DENCE IN (CONNECTIO	i with this	APPLICATION SHOULD	RE CENT
No Contra					210, Alexandria, Virginia 2	
Telephone: (703		IDEL, L.L.F.	., 1421 TIME	Succi, Suite	210, Alexandria, Virginia 2	2314-2003
reichfidue: (vo	3) 139-0220.					
I hereby de	clare that I have	reviewed and ur	nderstand the conte	nts of this Declarat	ion, and that all statements made here	in of my own
					e true; and further that these statemen	
					ne or imprisonment, or both, under So	
little 18 of the United	1 States Code and	d that such willful	i iaise statements m	ay jeopardize the vi	lidity of the application or any patent i	zzaca micreon.
3 Typewritten Full I Sole or First Inven		Junichi			HANNA	
		Giyer	n Name	Middle Initia		
		Gu	n-ichi		Hanna	
*4 Inventor's Signate	ure 🖙		11200			
5 Date of Signature	137	April		9,	1998	
3 24.0 0. 0.g		Month		Day	Year	
	Shinju	ıku-Ku	Tokyo-	То	Japan	
6 Residence					Country	
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8 Post Office	• Address	c/o Da	i Nippon	Printing Co	o., Ltd., 1-1, Ichiga	aya-Kaga
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address, in	cluding country)	$\frac{\text{cno } 1}{\text{cno } 1}$	Chome, Sni	njuku-Ku, '	Tokyo-To, Japan	

^{*}This form may be executed only when attached to the specification (including claims) at the end thereof if Box a. is checked.

^{**}Note to the Inventor. Please sign name on line 4 exactly as it appears in line 3 and insert the actual date of signing on line 5.

PAGE 2 OF U.S.A. DECLARATION FORM (Discar page in a sole inventor application)

'3 Typewritten Full Name		Masahir	0		FUNAHA	THP
Second Joint Inventor (if	any)		en,Name	Middle Initial		
		//	hiro	Widdle Hillar	Family Name	
*4 Inventor's Signature	(ar	or Cara			tunchashu	
5 Date of Signature	ar A	pril		9,	1998	
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6 Residence	Shin	juku-Ku	Toky	0-T0	Japan	
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3 Typewritten Full Name	of					
Third Joint Inventor (if a		Kom	ei		KAFUKU	
		Giv	en Name	Middle Initial	Family Name	
*4 Inventor's Signature	æ	Lon	nei		Labuhu	'n
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6 Residence		Las Vegas		evada	U.S.A.	
7 Citizenship	Ja	City pan	State or P	Tovince 	Country	
Post Office Addr (Insert complete address, including	mailing	8060 Cast	le Pines)	Avenue, Las V	⁷ egas, Nevada 89	113 U.S.A.
3 Typewritten Full Name Fourth Joint Inventor (if		Ку	roko		KOGO	
			en Name	Middle Initial	Family Name	
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6 Residence	Sninj	uku-Ku		yo-To	Japan	
7 Citizenship	Japan	City	State or F	rovince	Country	
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(Insert complete address, including	v	1-Chome, S	Shinjuku-K	u, Tokyo-To,	Japan	
3 Typewritten Full Name Fifth Joint Inventor (if a		Giv	ven Name	Middle Initial	Family Name	
*4 Inventor's Signature	a	···				
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6 Residence		Cin	C			
7 Citizenship		City	State or F	TOVINCE	Country	
8 Post Office Addr (Insert complete address, including	mailing g country)	Line 4		2 1 1	late of signing on line 5	

^{*}Note to Inventors: Please sign name on line 4 exactly as it appears in line 3 and insert the actual date of signing on line 5.

**This form may be executed only when attached to the first page of the Declaration and Power of Attorney form and the specification (including claims) of the application to which it pertains.